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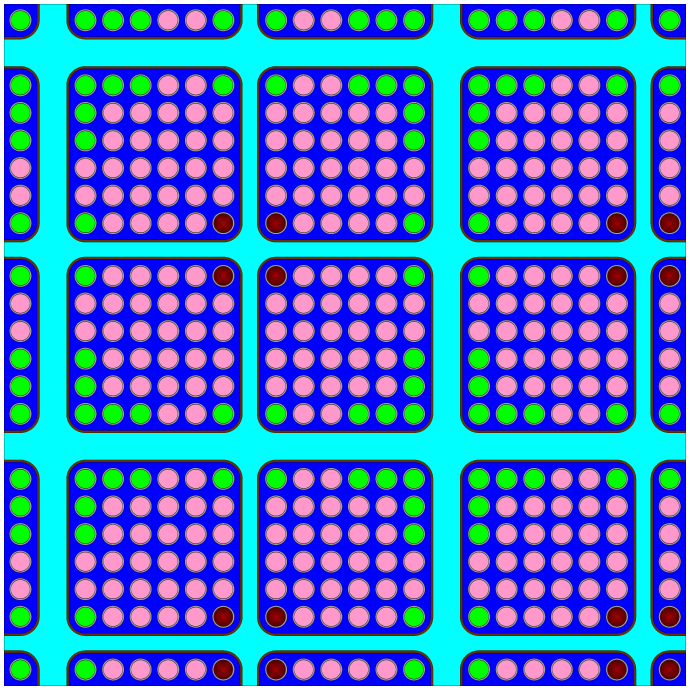
Published: 02/07/2021

Document Version
Publisher's final version

RESEARCH REPORT

VTT-R-00632-21



Sensitivity and uncertainty analysis of Gundremmingen-A assembly B23 sample I2680 depletion calculation with Serpent 2

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Confidentiality: Public

Report's title Sensitivity and uncertainty analysis of Gundremmingen-A assembly B23 sample I2680 depletion calculation with Serpent 2	
Customer, contact person, address H2020-Euratom-1.2	**Order reference** Grant agreement ID: 847593
Project name Spent Fuel Characterization and Evolution Until Disposal	**Project number/Short name** 128908/EU_EURAD
Author(s) Silja Häkkinen	**Pages** 21/409
Keywords SNF, burnup, nuclide inventory, Gundremmingen, SFCOMPO-2.0, benchmark, EURAD, sensitivity, uncertainty	**Report identification code** VTT-R-00632-21

Summary

The work described in this report presents sensitivity and uncertainty calculations in EU project EURAD work package 8 Spent Fuel Characterization and Evolution Until Disposal (SFC) subtask 2.1. Sensitivity and uncertainty analysis is performed in Serpent 2 depletion calculations of one sample in a 6x6 BWR assembly. Calculated sensitivites and uncertainties to decay heat and concentrations of several nuclides are presented. The Serpent calculated nuclide concentrations are compared to measured concentrations available in SFCOMPO-2.0.

The calculations were performed on a two dimensional assembly. Sensitivities and uncertainties on several operating history parameters, fuel properties and computational methods were calculated. Uncertainties in burnup were by far the most significant uncertainty component for decay heat and the studied nuclides ^{14}C, ^{36}Cl, ^{137}Cs, ^{148}Nd, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{242}Cm and ^{244}Cm. The only exception was ^{239}Pu that was most sensitive to water density (moderator density and void fraction). Other generally rather significant contributors to uncertainty were water density (moderator density and void fraction) and fuel density. Uncertainties in pin radius or ^{234}U enrichment had small or insignificant impact to the uncertainties of the calculated quantities. Uncertainties in decay data had some impact only on ^{242}Cm concentration and decay heat at 0 cooling time. The impact of the other studied uncertainty components, power density, water and fuel temperature, ^{235}U enrichment and ^{238}U content, were more dependent on the calculated quantity. According to the sensitivity studies fuel swelling, cross section data and fission yield data may have significant impact on many of the calculated quantities.

Confidentiality	Public

Espoo 2.7.2021

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Contents

1. Introduction

The work presented in this report is related to EU project EURAD (European Joint Programme on Radioactive Waste Management) work package number 8 SFC (Spent Fuel Characterization and Evolution Until Disposal). The work package consists of five tasks including i) coordination and training, ii) fuel characterization and related uncertainty analysis, iii) fuel and cladding behaviour and interaction after discharge, iv) accident scenario and consequence analysis and v) civil society interaction. This work is related to task 2 subtask 2.1.

The main objective of task 2 is to produce experimentally verified procedures to determine reliable source terms of spent nuclear fuel (SNF), including realistic uncertainties. The different subtasks include both computational and experimental spent fuel characterization and method development. Nuclide inventory of the fuel cladding is also investigated. Subtask 2.1 consists of benchmark calculations, sensitivity and uncertainty analysis and identification of the significant irradiation history parameters influencing the SNF properties. This report focuses on the sensitivity and uncertainty analysis. Results of the benchmark calculation are discussed in reference [1]. All calculations have been perfomed with Serpent 2 [2].

The sensitivity calculations comprise sensitivity to operating history, fuel properties, burnup induced changes and computational methods. Most of the operating history and fuel property parameters are also considered in the uncertainty analysis. The investigated spent fuel properties include decay heat and concentrations of the same nuclides as in the benchmark calculations [1] complemented with a couple of other nuclides. The other nuclides are impurity originating ^{14}C and ^{36}Cl and significant decay heat producers ^{90}Sr and ^{241}Am. These have been calculated for the same fuel sample number I2680 taken from Gundremmingen BWR reactor assembly B23 as in reference [1].

This report is structured as follows. The essential characteristics of the calculated assembly including geometrical data and description of the operating history are presented in Section 2. Section 3 describes the calculation models in the reference calculation, the sensitivity calculations and the uncertainty analysis. Section 4 presents the results of all calculations and the results are discussed and summarized in section 5.

2. Fuel assembly B23 irradiated in Gundremmingen A

The calculations are based on a 6x6 BWR assembly B23 irradiated in the German reactor Gundremmingen A between 25.8.1969 – 5.5.1973. The assembly characteristics and irradiation history have been taken from SFCOMPO-2.0 [3]. More information on the reactor, SFCOMPO data and irradiation history is given in the benchmark report [1]. Only the essential characteristics necessary to explain the sensitivity and uncertainty calculations are given here.

Assembly B23 is depicted in Figure 1 [3]. The assembly includes two different fuel types with ^{235}U enrichments of 1.87 % (fuel 1) and 2.53 % (fuel 2). Rod A1, sample I2680 calculated in

the benchmark report [1], is of fuel type 2 and is presented in green colour in Figure 1. position of the cruciform control rod is indicated by the black lines in Figure 1.

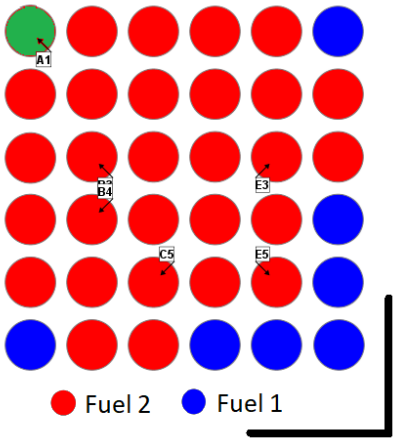

Figure 1. Modelled assembly B23 [3]. The calculated sample is presented in green colour.

The assembly and fuel rod dimensions are given in Table 1 [3–5]. The fuel density is 10.5 g/cm^3 [4]. Fuel cladding is composed of Zircaloy-2 [4, 5] and the channel wall of Zircaloy-4 [3–5]. The absorber material of the cruciform control rods is B_4C powder and the cladding material SS 304 [3–5].

Table 1. Assembly and fuel rod dimensions for the Gundremmingen A assembly B23 [3–5].

Assembly		Fuel rod	
Parameter	Value [cm]	Parameter	Value [cm]
Channel outer diam	11.352	Pin pitch	1.78
Channel inner diam	11.052	Fuel diam	1.224
Assembly pitch wide	13.098	Gas gap	0.01375
Assembly pitch narrow	12.303	Clad thickness	0.0889
Active length	330.2	Pin diam	1.428

The irradiation history of the assembly B23 is presented in Tabel 2 and some operating history parameters are given in Table 3 [3–5]. There is no information on the control rod operating history.

Table 2. Irradiation history of the Gundremmingen A assembly B23 [3–5].

Cycle of operation	Time period	Duration [EFPD]	Burnup increment [MWd/kgU]
Second	25.8.69-30.5.70	279	5.839
Shut down	31.5.70-24.7.70	56	
Third	25.7.70-12.6.71	323	6.131
Shut down	13.6.71-15.7.71	33	
Fourth	16.7.71-30.4.72	290	5.483
Shut down	1.5.72-30.6.72	61	
Fifth	1.7.72-5.5.73	309	5.174

Table 3. Operating history parameters of the Gundremmingen A assembly B23 [3–5].

Parameter	Value	Unit
Void at 268 cm	50	%
Void at 44 cm	0	%
Coolant inlet temp	539	K
Coolant outlet temp	559	K
Fuel temp	923	K
Coolant pressure	69	bar

3. Models and methods

All calculations were performed using Serpent 2, the continuous-energy Monte Carlo particle transport code, developed at VTT [2]. This chapter describes the Serpent model used in reference calculations, in the different parameter variations applied in the sensitivity calculations and in the uncertainty calculations.

3.1 Reference calculations

The Serpent model in the reference calculation was almost the same as that described in reference [1]. The parts that differ are typed in bold text.

The two dimensional Serpent model applied in the reference calculations is presented in Figure 2. Sample I2680 is indicated with dark red colour. Pink colour is used for the other fuel pins of the same type with 2.53 % ^{235}U enrichment. The green pins are type 1 with ^{235}U enrichment 1.87 %. Dark blue indicates coolant and light blue moderator. The used references did not include information on the rounded corners of the channel walls although e.g. figure 1.33 in reference [5] clearly indicate that the corners are rounded. The radius of the rounded corners was calculated so that the distance from the flow channel outer wall to the fuel pin surface remained constant. Thus, the inner and outer radius of the rounded corners in the model were 1.076 cm and 1.226 cm, respectively.

The 180° symmetry of the assembly was utilized in the Serpent model. The fuel was divided into pin-wise material zones using Serpent's automated depletion zone division. Additionally, sample I2680 was divided in ten equal size radial depletion zones. Material volumes were defined using the "set mvol" card.

No information was given on the ^{234}U content in the fuel, so it was omitted in the reference calculations. No information on impurities was given in the specifications. **In order to see the effect of the parameter variations on impurites, 10 ppm of ^{14}N and ^{35}Cl were added in the fuel**. These nuclides were selected because their activation products ^{14}C and ^{36}Cl are the most significant impurity originated mobile nuclides in spent nuclear fuel [6]. Altogether, the fuel in the model contained ^{235}U, ^{238}U, ^{16}O, ^{14}N and ^{35}Cl. Oxygen making 11.85 wt-% of the fuel. Zircaloy-2 and Zircaloy-4 compositions were taken from reference [7]. The elemental compositions given in the reference were decomposed into isotopic compositions using Serpent's "-elem" command option. The elemental compositions from [7] are presented in Table 4.

The reference calculations were normalized to power density at sample I2680 position, 22.814 W/gU, calculated from the effective power days and sample I2680 burnup. The sample burnup, 27.4 MWd/kgU, was based on Nd-148 measurements.

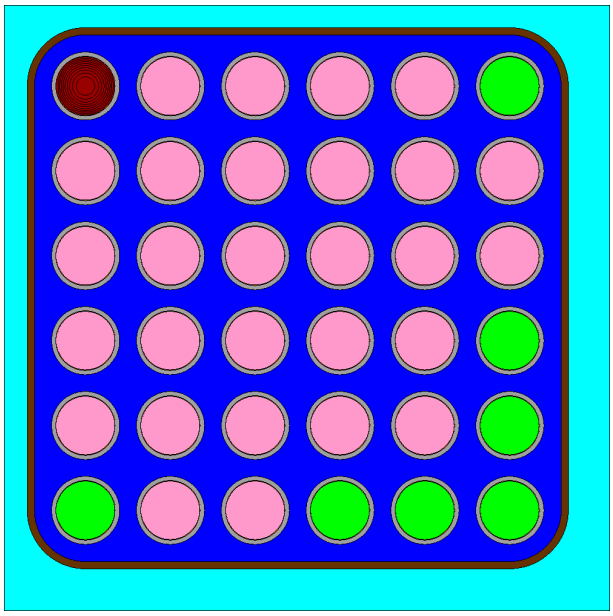

Figure 2. Two dimensional Serpent model of the Gundremmingen A assembly B23. The dark red fuel pin is sample I2680.

Table 4. Elemental compositions of Zry-2 and Zry-4 in the Serpent calculations.

Element	Zircaloy-2 [%]	Zircaloy-4 [%]
Sn	1.5	1.5
Fe	0.15	0.2
Cr	0.1	0.1
Ni	0.05	-
Zr	98.2	98.2

As for other operating history data, fuel temperature was assumed constant at the value 923 K throughout the calculation. Coolant and moderator temperatures were estimated as the average of inlet and outlet temperatures yielding 549 K. Additionally Serpent needs information on water densities. These were obtained by creating a CASMO-4E model [8] of the assembly and making a calculation in 69 bar pressure and 50 % void. The coolant and moderator densities could then be obtained from the CASMO output file. Information on control rod movements was not available so they were assumed to be fully withdrawn during the whole operation and were omitted from the model.

Altogether 69 burnup steps plus three decay steps between cycles were applied to irradiate the fuel for 1201 effective power days plus 150 days of shutdown between the cycles. Each cycle was started with a depletion step of 3 days which corresponds to less than 0.1 MWd/kgU. The step length was gradually incresed to 20 days. The length of the step was never more than double the previous step length. In two of the irradiation cycles, the last step was longer, 26 or 30 days.

The reference calculations were run with JEFF based nuclear data. Cross sections based on JEFF-3.2 and fission yield and radioactive decay data based on JEFF-3.1.1 were used. Thermal scattering data for light water was based on JEFF-3.1.

1E4 neutrons were modelled in every 200 active cycles and 30 inactive cycles. The sufficiency of the neutron histories was checked by examining the Shannon entropy of the source

point distribution. The assembly was modelled in an infinite lattice using reflective boundary conditions. The substep method developed for Serpent [9] was used in the burnup calculation applying linear extrapolation in predictor and linear interpolation in corrector with 10 substep in both predictor and corrector. Doppler-broadening rejection correction was used for some uranium and plutonium nuclides. The reference calculations were repeated ten times with different random number generator seed values to get an idea of the variance caused by the Monte Carlo method on the results. The average of the ten calculations was taken as the reference result.

3.2 Sensitivity calculations

The sensitivity of specified SNF properties on operating history parameters, fuel properties and computational methods have been studied by varying the parameters and examining the changes in the SNF properties. The varied parameters are presented in Table 5. When one parameter was varied all the other input parameters in the Serpent model were kept as in the reference calculation except for those that are directly affected by the change in one parameter such as e.g. oxygen content when varying ^{238}U content.

In the reference calculations, burnup steps are defined as effective power days (EFPD) and the calculation is normalized to power density. This means that if power density is varied, also burnup changes. In order to see the effect of power variations with constant burnup, the burnup steps were defined in MWd/kgU in variation calculations POW1 and POW2. For comparison, another reference calculation was also conducted using burnup steps in MWd/kgU. In these calculations assembly burnup of each cycle from [5] was used and the calculation was normalized to assembly power density (18.84 W/gU) calculated from the assembly average burnup (22.627 MWd/kgU [5]) and effective power days.

Fuel swelling has been examined by increasing the fuel pellet radius step by step and proportionally decreasing fuel density so that fuel mass remains constant. The radius given in Serpent's "div" card for depletion zone division and the material volumes defined by Serpent's "set mvol" card were also increased accordingly. This has been realized by dividing each cycle in two except for the last cycle. The first half of the first cycle applied the nominal fuel pellet radius and after that the radius was increased at every half cycle. The last cycle was divided in two approximately equal parts and a third part that included tha last depletion step of 20 EFPD. The largest fuel pellet radius was applied only for the last step.

The ranges and reference values of the varied parameters are listed in Table 6. In case of HIS1 and HIS2, the number reflects how many EFPDs have been added or subtracted from the total number of EFPDs. In case of SWE, the number reflects how many percentages of the original gap width remains, i.e. 0 means that at the end of irradiation the fuel is in contact with the cladding and 90 means that the gap width is 90 % of the original gap width. For DBRC, the nuclides included in doppler broadening rejection correction and the minimum and maximum energies are given. In the variation "energy", only the energy range has been changed. In nuclear data library variations, the thermal scattering data for light water from JEFF-3.1.1 was always used. The abbreviations of the time integration methods applied in the substep variations are listed below.

- LELI – Linear extrapolation in predictor and linear interpolation in corrector
- CECE – Constant extrapolation in predictor and constant backwards extrapolation in corrector
- CELI – Constant extrapolation in predictor and linear extrapolation in corrector
- LEQI – Linear extrapolation in predictor and quadratic interpolation in corrector

Table 5. Varied parameters in the sensitivity calculations. The dashed lines indicate a change in the type of varied parameters: operating history, fuel properties and computational methods.

DMOD	Moderator density
HIS1	Variation in EFPD. Variations have been divided equally between all cycles. I.e. an addition of 12 EFPD means that the length of each cycle has been increased by 3 EFPD.
HIS2	Like HIS2 but all variations have been added to the last cycle keeping the other cycles' lengths in their reference values.
POW1	Power density variations with constant burnup.
POW2	Like POW1 but the last of the four cycles with reference power density
POW_BU	Power density variations with constant irradiation time (burnup changes)
TFUE	Fuel temperature, all fuel pins in the assembly
TFUE_1	Fuel temperature, only sample I2680
TWAT	Water (coolant and moderator) temperarure
VOID	Void fraction (variations in coolant density)
DFUE	Fuel density, all fuel pins
DFUE_1	Fuel density, only sample I2680
PIN	Pin radius, all fuel pins
PIN_1	Pin radius, only sample I2680
SWE	Fuel swelling, all fuel pins
U234	^{234}U enrichment (percentage of ^{235}U), all fuel pins
U234_1	^{234}U enrichment (percentage of ^{235}U), only sample I2680
U235	Assembly average ^{235}U enrichment
U235_1	^{235}U enrichment of sample I2680
U238	Average ^{238}U content in the assembly. ^{235}U mass fraction in the fuel is the same as in reference. Oxygen content varies with varying ^{238}U.
U238_1	^{238}U content in sample I2680
DBRC	Doppler broadening rejection correction
LIB	Nuclear data library
SUB	Substep method, time integration mode

3.3 Uncertainty analysis

The uncertainty analysis was conducted on the studied sample I2680 of the Gundremmingen assembly B23. The analysis is based on random sampling of parameter values from a normal distribution. Each studied parameter was randomly sampled around a given mean (used in the reference calculation) with a given standard deviation and a burnup calculation was conducted with each varied parameter. The variance of the calculated SNF property $\sigma^2_{\text{tot},i}$ reflects the variance caused by the sampled input parameter σ^2_i and the variance from the Monte Carlo method σ^2_{MC} according to equation 3.1.

$$\sigma^2_{\text{tot},i} = \sigma^2_i + \sigma^2_{\text{MC}}. \tag{3.1}$$

The value for σ^2_{MC} was calculated by repeating the reference calculation 200 times. The random sampling of the studied input parameters was repeated 200 – 250 times depending on the calculated parameter. The sufficiency of the number of sampling calculations was confirmed by

Table 6. Parameter ranges in the sensitivity calculations.

Parameter	Unit	Nominal	Lower limit	Upper limit	Step	# of var
DMOD	g/cm^3	0.759	0.659	0.859	0.02	10
HIS1/2	EFPD	0	-20	20	4	10
POW1/2	W/gU	18.84	10.0	30.0	1.0 / 6.0 / 4.0	11
POW_BU	W/gU	22.814	16.0	26.0	1.0	10
TFUE	K	923	863	983	5 / 10	20
TFUE_1	K	923	898	948	5	10
TWAT	K	549	509	589	2 / 5 / 10	20
VOID	%	50	0	100	2 / 20 / 40	13
DFUE(_1)	g/cm^3	10.5	10.0	11.0	0.1	10
PIN(_1)	cm	0.612	0.598	0.625099	0.02	14
SWE	%	100	0	90	10	10
U234(_1)	%	0	0.1	1.0	0.02 / 0.1	18
U235	%	2.40	2.30	2.50	0.01 / 0.05	12
U235_1	%	2.53	2.43	2.63	0.01 / 0.05	12
U238	%	86.03	85.93	86.13	0.02	10
U238_1	%	85.92	85.82	86.02	0.02	10

Parameter	Reference	Variations	# of var
DBRC	^{234}U ^{235}U ^{238}U ^{239}Pu ^{240}Pu 1E-7 - 1E-3	all; actinides (^{233}U – ^{246}Cm); energy (1E-8 – 1E-2); no DBRC	4
LIB	JEFF-3.20 (XS) JEFF-3.1.1 (FY, DD)	ENDF/B-VI (no dbrc); JENDL-4.0; ENDF/B-VII.1	3
SUB	LELI	CECE; CELI; LEQI; no substeps	4

requiring that the calculated variance had converged and more calculation cases would keep the changes in the variance approximately within 2 % (i.e. $\pm$1 %). This check was performed for the variance in sample decay heat and ^{238}Pu concentration and it was assumed to hold for the other nuclide concentrations as well. The sufficiency of the repetitions of the reference calculation was checked similarly. For the reference calculation, convergence was reached with $\sim$ 100 repetitions. The high number of reference calculations needed arises from the rather small number of neutron histories, altogether 2E6, used in the calculations. Increasing the number of neutron histories would likely decrease the Monte Carlo variations, but would also increase the calculation time. The total variance on a given SNF property was then calculated by equation 3.2

$$\sigma_k^2 = \sum_{i=1}^{n} \sigma_i^2, \tag{3.2}$$

where k refers to the SNF property such as e.g. decay heat or ^{238}Pu concentration and i refers to an input parameter such as e.g. fuel temperature or moderator density.

It should be noted that quation 3.2 is essentially an approximation of the total variance of a given SNF property and the square root of equation 3.2 is an approximation of the total uncertainty. This is because all of the parameters σ_i are not completely uncorrelated, but some correlations

may exist. For example water temperature and water density are not independent of each other and fuel temperature effects water temperature. However, for the purposes of this study the approximative uncertainty estimate is considered sufficient.

The different input parameters *i* used in the uncertainty calculations, their mean values and standard deviations are presented in Table 7. The parameter acronyms are the same ones explained in Table 5. The additional acronyms BU and DEC refer to variations in burnup and decay data. No specified mean or standard deviation values are given to the decay data. Its effect on the SNF properties has been examined by random sampling the uncertainties found in the ENDF-6 format nuclear data from log-normal distribution similarly to references [10, 11].

Table 7. Input parameters sampled in the uncertainty calculations. Mean values of the parameters (Mean), standard deviations (Stdev), relative standard deviation with respect to mean (Rsd) and units of Mean and Stdev.

Parameter	Mean	Stdev	Rsd [%]	Unit
DMOD	0.759	0.015	2	g/cm^3
POW1	18.84	0.31	1.7	W/gU
TFUE_1	923	18	2	K
TWAT	549	5.49	1	K
VOID$_1$	50	3.3	6.7	%
VOID$_2$	50	0.67	1.3	%
DFUE_1	10.5	0.03	0.3	g/cm^3
PIN_1	0.612	0.0002	0.04	cm
U234_1	0.65	0.08	0.1	%
U235_1	2.53	0.017	0.7	%
U238_1	85.92	0.57	0.7	%
BU	22.627	0.453	2	MWd/kgU
DEC	–	–	–	–

The mean values in Table 7 are the same that have been used in reference calculations described in section 3.1 except for ^{234}U concentration which was zero in the reference calculations explained in section 3.1. Another set of reference calculations were conducted for the uncertainty calculations using the mean value for ^{234}U enrichment in ^{235}U presented in Table 7. A non-zero mean for ^{234}U was used in order to sample the ^{234}U enrichment from normal distribution like for the other parameters.

Accurate estimations for the standard deviations of the input parameters should ideally be derived from measured time dependent irradiation history data and manufacturing tolerances. However, such information was not available and other sources of information were used to evaluate estimates for the standard deviations. Standard deviation for the moderator density (DMOD) has been estimated from the standard deviation of known irradiation history data for a VVER-440 reactor. Standard deviations for power density (POW1) and water temperature (TWAT) have been taken the same as given for a BWR reactor in the UAM II benchmark specification (Table 2) [12]. Standard deviations for fuel density (DFUE_1) and fuel radius (PIN_1) are taken to be the same as given in UAM I benchmark specification (p. 40) for a BWR assembly [13]. Standard deviation for ^{235}U enrichment (U235_1) is based on the tolerances ($\pm$0.05 %) given in the UAM I benchmark for a VVER-1000 assembly (p. 40) [13]. The standard deviation has been calculated as one third of the upper and lower bounds following the example of [12]. Standard deviation for void (VOID) and ^{234}U enrichment (U234_1) in ^{235}U are also calculated as one third of upper and lower limits. For ^{234}U, the upper and lower limits (0.4 and 0.9 % in ^{235}U) are values found in SFCOMPO for those measured BWR samples that gave

a value for ^{234}U. The mean value for ^{234}U given in Table 7 is the average of these limits. For void the limits are purely a guess and therefore two sets of upper and lower limits were used, $\pm$10 % and $\pm$2 %. The standard deviation for ^{238}U (U238_1) is based on assuming the same relative standard deviation as for ^{235}U. The standard deviation for fuel temperature (TFUE_1) is an educated guess. Standard deviation for burnup is the measurement uncertainty reported in SFCOMPO-2.0 for sample burnup from ^{148}Nd measurement.

In addition to the parameters listed in Table 7, an uncertainty estimate related to DBRC was also derived. This was done by comparing the results of the reference calculation to a calculation applying DBRC on all nuclides. An estimate on the uncertainty caused by not using DBRC was also evaluated by comparing a calculation not applying DBRC to the calculation applying DBRC on all nuclides. The calculations with DBRC on for all nuclides and DBRC off were repeated 110 times ans the average of the 110 calculations were used for the comparison. The repetitions were made in order to insure that the calculation has converged.

4. Results

Appendix A presents figures of decay heat and nuclide concentrations with different parameter variations (see Table 6). The nuclides included in the examination are those 12 nuclides whose measured concentrations are found in SFCOMPO-2.0 [3] for Gundremmingen assembly B23 sample I2680 and are presented in the benchmark report [1]. Additionaly ^{14}C and ^{36}Cl have been included as significant impurity originating nuclides and ^{90}Sr and ^{241}Am have been included due to their contribution to decay heat.

For each parameter and decay heat or nuclide concentration, two figures are presented. The first figure presents the relative difference of calculated decay heat or nuclide concentration with all parameter variations to the reference calculation as a function of cooling time. The relative standard deviation of the repetition calculations of the reference, denoted as "std" in the figures, is also presented. The absolute value of the reference calculation is presented on the right hand side axis. The horizontal axis in the figures is in logarithmic scale and hence does not start from 0 but from 1 day.

The second figure presents on the left hand side axis the calculated decay heat or nuclide concentration as a function of parameter variation including a linear fit to the calculated points. The linear fit has been done using Matlab's polyfit function which uses the Least Squares Method. The right hand side axis presents the relative difference to the reference and for the reference calculation the relative standard deviation of the repetitions.

Table 8 summarises the results of the figures in Appendix A at 0 cooling time. The Table presents the percentual change in the calculated decay heat or nuclide concentration with 2 % change in the varied parameter based on the linear fit to the calculated results. In case the changes in the nuclide concentration or decay heat are insignificant within the parameter range calculated or clearly nonlinear, a dash is presented. Insignificance is estimated from the figures in Appendix A and the standard deviation of the repetitions of the reference calculation. If the change in nuclide concentration or decay heat is not significant within the 2 % change in parameter variation, this is indicated by enclosing the number in brackets. The purpose of the Table is to visualize the sensitivity of each calculated quantity to different input parameters. The changes in the Table are coloured red when feedback to parameter variation is positive and blue when it is negative.

*Table 8. Change in decay heat and nuclide concentrations [%] with 2 % change in the input parameter. Positive feedback is presented with red and negative feedback with blue colour. Insignificance of a parameter deviation within the calculated parameter range (Table 6) is indicated with a dash. * Numbers give the difference to reference when fuel rod gap size is zero at the end of irradiation. The brackets indicate that the change in decay heat or nuclide concentration is not significant within the 2 % change in the input parameter.*

	DH	^{14}C	^{36}Cl	^{90}Sr	^{137}Cs	^{148}Nd	^{235}U	^{236}U	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{242}Cm	^{244}Cm
DMOD	–	0.42	0.51	0.12	–	(0.002)	-0.65	0.06	0.01	-0.86	-1.0	-0.19	-0.85	-0.07	-1.1	-0.63	-1.2
HIS1	–	1.1	1.0	0.8	1.0	1.0	5.1	0.58	-0.05	1.4	0.25	1.0	1.0	1.4	1.1	1.4	1.6
HIS2	–	1.1	1.0	0.8	1.0	1.0	5.1	0.58	-0.05	1.4	0.28	1.0	1.0	1.4	1.0	1.3	1.6
POW1	2.0	–	–	0.09	0.0	0.03	–	–	(0.000)	-0.50	–	–	0.08	(0.03)	-1.7	-0.47	0.06
POW2	–	–	–	0.06	0.05	0.02	–	–	(0.000)	-0.28	0.02	–	0.03	–	-0.51	-0.80	(0.05)
POW_BU	2.0	2.2	2.0	1.6	2.0	2.0	-3.6	0.98	-0.051	4.0	0.37	1.9	2.1	4.1	1.1	3.5	6.7
TFUE	–	-0.10	-0.10	-0.03	–	–	0.15	-0.05	-0.002	–	0.16	–	0.23	0.11	0.30	0.17	–
TFUE_1	–	-0.08	-0.08	-0.02	–	–	0.13	–	-0.002	–	0.12	–	–	–	0.24	–	–
TWAT	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
VOID	0.02	-0.30	-0.39	-0.09	(-0.001)	(-0.002)	0.49	-0.03	-0.01	0.62	0.8	0.21	0.61	–	0.82	0.41	0.92
DFUE	2.0	1.6	1.5	1.9	2.0	2.0	2.6	2.0	2.0	3.1	2.9	2.3	2.5	1.8	2.8	2.4	3.7
DFUE_1	2.0	2.0	1.9	2.0	2.0	2.0	2.1	2.0	2.0	2.4	2.0	2.2	1.8	1.7	1.8	1.9	2.6
PIN	4.0	-1.0	-1.3	-0.28	–	–	1.6	(-0.09)	-0.02	2.3	2.2	0.66	1.8	–	2.3	1.4	3.7
PIN_1	(4.0)	-0.14	-0.25	-0.05	(-0.009)	–	0.30	–	0.00	0.80	0.29	–	–	–	–	–	1.1
SWE*	0.2	-2.5	-2.4	-1.6	-1.9	-1.9	3.7	0.90	0.03	-2.5	1.5	-1.5	-0.67	-4.6	1.7	-3.0	-4.2
U234	–	-0.004	-0.004	0.001	–	–	0.018	0.003	0.00	–	0.003	–	–	-0.007	–	–	–
U234_1	–	-0.003	-0.003	0.001	–	–	0.017	0.004	0.00	–	0.001	–	–	–	–	–	–
U235	–	-1.4	-1.43	0.60	-0.008	-0.01	4.1	1.4	-0.03	-0.79	0.27	-0.91	-0.48	-2.2	–	-1.4	-4.1
U235_1	–	-1.3	-1.28	0.68	-0.01	-0.02	4.0	1.5	-0.03	-1.1	–	-0.91	-0.92	-2.5	-0.57	-1.9	-4.9
U238	–	1.3	0.90	1.3	2.0	2.1	-1.7	–	2.0	–	2.1	–	–	–	–	–	–
U238_1	–	1.6	1.34	1.3	1.9	1.9	-2.3	–	2.0	–	–	–	–	6.2	–	–	–

For swelling a 2 % change in the gap size is not very informative, since swelling is typically much stronger than just 2 % of the gap size. Therefore the values presented for swelling in Table 8 are the differences to reference calculation when 0 % of the gap remains at the end of irradiation. For fuel temperature, pin radius and ^{234}U enrichment, the 2 % change in parameter value applied in Table 8 is unrealistically large compared to the values estimated in Table 7.

Sensitivity to water temperature and in many cases to fuel temperature, ^{238}U content and ^{234}U enrichment is clearly non-linear. Nuclides heavier than uranium seem to have mostly negative feedback on moderator density and ^{235}U concentration and positive feedback on changes in irradiation time, burnup, void fraction, fuel density and pin radius. Naturally, all nuclides and decay heat have a positive feedback on fuel density.

According to the results in Table 8, the sensitivity to irradiation days (HIS1 and HIS2) doesn't depend on whether the change in irradiation days happens at the end of irradiation (HIS2) or is evenly distributed among the cycles. Power density variations only in the first three cycles (POW2) don't seem to have an effect on decay heat, but are clearly important if the variations are extended to the last cycle.

Calculational methods such as usage of DBRC and the substep method have rather small effects on the results. DBRC has practically no effect on decay heat at zero cooling time. For the nuclide concentrations its effect is less than 1 % except for ^{241}Am that has a $\sim$ 1 % change to reference if DBRC is not used. Mostly there is no effect at all unless DBRC is off. Even then the effect on the fission products ^{90}Sr, ^{137}Cs and ^{148}Nd are less than 0.1 %.

Nuclides ^{239}Pu, ^{236}U and ^{241}Pu did not react to the time integration method. Most differences to reference in the other nuclides with different time integration methods occur when constant extrapolation in predictor and constant backwards extrapolation in corrector (cece) is used. This method caused larger differences to reference than not using substeps at all. Decay heat exhibited minor $\sim$ 0.1 % differences when substeps were not applied. All differences to reference were below 0.9 % except for ^{244}Cm $\sim$ 1.5 %.

Differences to reference due to different nuclear data were clearly larger than differences in DBRC or substep method. The largest differences due to nuclear data and the library responsible for the largest difference are presented in Table 9. The largest differences occur mostly with the ENDF/B-VI.8 nuclear data which is not surprising since it is the oldest library used compared to the reference nuclear data JEFF-3.2. Significant differences of several percents occur for ^{14}C, ^{36}Cl, ^{238}Pu, ^{241}Am, ^{242}Cm and ^{244}Cm. The differences are more than 1 % also for decay heat and the other nuclides except ^{137}Cs, ^{236}U, ^{238}U and ^{242}Pu. Differences in ^{238}U concentration between the different nuclear data are insignificant.

Table 10 presents the calculated uncertainties from different input parameters on sample I2680 decay heat and nuclide concentrations at 0 cooling time. The total uncertainties ΔC_1 and ΔC_2 are relative standard deviations based on equation 3.2. The subindexes refer to total uncertainty calculated with the different void fraction uncertainties. $DBRC_{on}$ gives the percentual difference of reference calculation to a calculation where DBRC has been applied to all nuclides. $DBRC_{off}$ gives the percentual difference of a calculation where DBRC was not used to the calculation where DBRC was used for all nuclides. The numbers in the brackets mean that the standard deviation of the reference repetition calculations are larger than the difference given in the brackets. The DBRC results have not been included in the calculation of total uncertainty ΔC_i.

Table 11 presents the calculated and measured nuclide concentrations and their ratios reported in [1]. The Table also includes the measurement uncertainties reported in SFCOMPO-2.0

Table 9. Largest differences to reference (JEFF-3.2) due to nuclear data. Column "Library" gives the nuclear data library that exhibits the largest difference.

Nuclide	Library	Difference [%]
DH	ENDF/B-VII.1	1.5
^{14}C	ENDF/B-VI.8	6.5
^{36}Cl	ENDF/B-VI.8	5.4
^{90}Sr	ENDF/B-VII.1	1.7
^{137}Cs	ENDF/B-VII.1	0.5
^{148}Nd	ENDF/B-VI.8	1.2
^{235}U	ENDF/B-VI.8	1.2
^{236}U	ENDF/B-VII.1	0.4
^{238}U	ENDF/B-VI.8	0.002
^{238}Pu	ENDF/B-VI.8	6.1
^{239}Pu	ENDF/B-VI.8	1.1
^{240}Pu	JENDL-4.0	1.3
^{241}Pu	ENDF/B-VI.8	1.5
^{242}Pu	ENDF/B-VI.8	0.9
^{241}Am	ENDF/B-VI.8	20
^{242}Cm	ENDF/B-VI.8	10
^{244}Cm	ENDF/B-VII.1	11

Table 10. Input parameter uncertainties and total uncertainties (ΔC) on decay heat and nuclide concentrations [%] at 0 cooling time. $DBRC_{on}$ gives the difference to reference when DBRC has been used for all nuclides and $DBRC_{off}$ gives the difference between DBRC off and DBRC used for all nuclides. The numbers in the brackets mean that the relative difference is smaller than the standard deviation of the reference repetition calculations. DBRC has not been included in the total uncertainty ΔC.

	DMOD	POW1	TFUE_1	TWAT	$VOID_1$	$VOID_2$	DFUE_1	PIN_1	U234_1	U235_1	U238_1	BU	DEC	$DBRC_{on}$	$DBRC_{off}$	$\Delta\mathbf{C_1}$	$\Delta\mathbf{C_2}$
DH	0.00	1.79	0.00	0.00	0.12	0.07	0.27	0.00	0.00	0.24	0.21	0.03	0.32	(0.04)	(0.05)	1.85	1.85
^{14}C	0.51	0.00	0.04	0.14	0.93	0.21	0.25	0.01	0.02	0.09	0.06	2.09	0.00	(0.01)	0.19	2.37	2.19
^{36}Cl	0.60	0.00	0.05	0.15	1.19	0.26	0.25	0.01	0.02	0.12	0.07	1.89	0.00	(0.01)	0.21	2.33	2.02
^{137}Cs	0.12	0.07	0.04	0.04	0.06	0.02	0.27	0.01	0.00	0.30	0.11	1.78	0.00	(0.01)	(0.03)	1.83	1.83
^{148}Nd	0.12	0.03	0.04	0.04	0.06	0.02	0.27	0.01	0.00	0.30	0.12	1.82	0.00	(0.01)	(0.02)	1.87	1.87
^{235}U	0.72	0.00	0.07	0.24	1.37	0.30	0.51	0.02	0.07	0.82	0.09	2.75	0.00	(0.01)	0.29	3.32	3.04
^{236}U	0.11	0.00	0.02	0.02	0.16	0.03	0.32	0.00	0.02	0.63	0.03	0.82	0.00	(0.02)	0.11	1.10	1.09
^{238}U	0.01	0.00	0.00	0.00	0.02	0.00	0.36	0.00	0.00	0.02	0.68	0.04	0.00	(0.00)	0.01	0.77	0.77
^{238}Pu	0.58	0.40	0.00	0.00	1.75	0.34	0.22	0.00	0.00	0.27	0.16	4.53	0.00	(0.04)	(0.04)	4.92	4.60
^{239}Pu	0.88	0.01	0.16	0.39	2.12	0.46	0.34	0.00	0.00	0.02	0.40	0.29	0.00	(0.02)	0.56	2.41	1.23
^{240}Pu	0.05	0.00	0.07	0.21	0.51	0.09	0.30	0.00	0.00	0.02	0.44	1.73	0.00	(0.04)	(0.11)	1.89	1.82
^{241}Pu	0.67	0.07	0.23	0.07	1.70	0.36	0.23	0.00	0.00	0.00	0.17	1.82	0.00	(0.02)	0.53	2.60	2.00
^{242}Pu	0.18	0.07	0.20	0.32	0.04	0.08	0.12	0.00	0.07	0.13	0.07	4.23	0.02	(0.03)	0.20	4.26	4.26
^{242}Cm	0.38	0.42	0.23	0.25	1.18	0.24	0.17	0.04	0.00	0.05	0.11	4.02	0.28	(0.01)	0.37	4.26	4.10
^{244}Cm	0.77	0.00	0.16	0.38	2.56	0.51	0.00	0.00	0.00	0.26	0.00	8.70	0.01	(0.03)	(0.12)	9.12	8.77

and the calculated uncertainties from Table 10. Figure 3 presents the difference between calculated and measured nuclide concentrations (calculated/measured - 1)·100 for both 2D and 3D assembly calculations (similar to Figure 9 in [1]) and the combined uncertainties for the calculations and measurements ($\Delta C + \Delta E$). The smaller uncertainty value for void fraction has been used in the calculated uncertainty (C_2 in Table 11).

Table 11. Measured (E) and calculated (C) nuclide concentrations [g/kgUi] of sample I2680 with associated uncertainties (ΔE, ΔC_i) . The subindexes i refer to total uncertainties calculated with larger and smaller assumed uncertainty for void fraction.

Nuclide	E	C	C/E	ΔE [%]	ΔC_1 [%]	ΔC_2 [%]
^{137}Cs	0.915	0.994	1.09	1.5	1.8	1.8
^{148}Nd	0.309	0.306	0.99	1.0	1.9	1.9
^{235}U	6.74	6.19	0.92	1.0	3.3	3.0
^{236}U	3.26	3.08	0.94	1.2	1.1	1.1
^{238}U	952.3	953.7	1.00	2.1	0.77	0.77
^{238}Pu	0.108	0.097	0.90	2.0	4.9	4.6
^{239}Pu	4.80	4.62	0.96	0.88	2.4	1.2
^{240}Pu	2.17	2.12	0.98	1.0	1.9	1.8
^{241}Pu	1.14	1.02	0.89	1.2	2.6	2.0
^{242}Pu	0.450	0.411	0.91	1.1	4.3	4.3
^{242}Cm	0.0146	0.0135	0.92	4.2	4.3	4.1
^{244}Cm	0.0198	0.0172	0.87	2.8	9.1	8.8

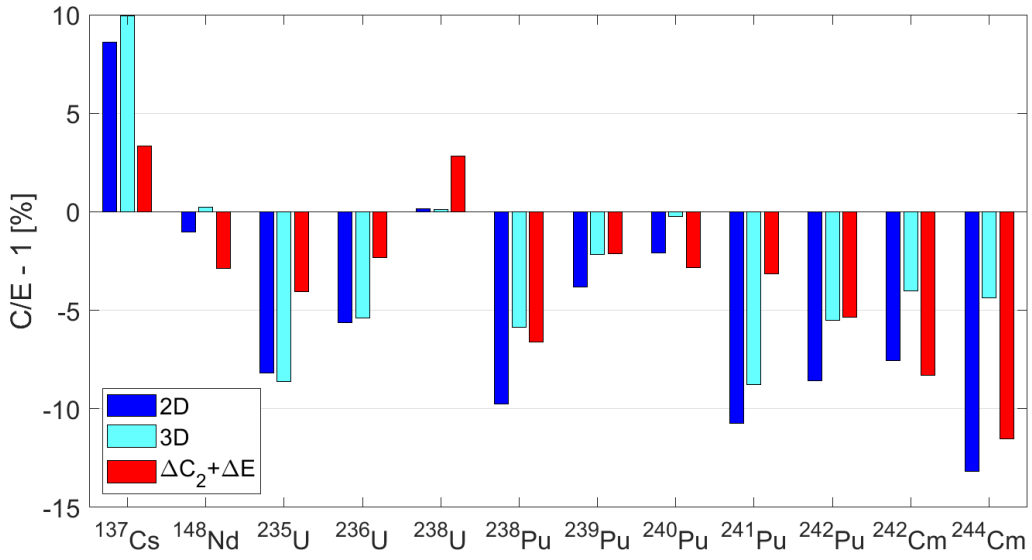

Figure 3. Calculated/Measured - 1 isotopic concentrations in % in the 2D and 3D calculations. Combined uncertainties from calculations and measurements are presented in red bars. Smaller uncertainty for void fraction was used in the calculation uncertainty.

The calculated results agree with the measurement results within the combined uncertainty for nuclides ^{148}Nd, ^{238}U, ^{240}Pu and ^{242}Cm for both 2D and 3D calculations. Additionally, the 3D calculation agrees with the measurement within these uncertainties for ^{238}Pu and ^{244}Cm. If the larger uncertainty for void fraction was used the 3D calculation of ^{239}Pu would also agree with the measurements within the combined uncertainty.

Significant uncertainty in the calculations, not considered in Tables 10 and 11, may arise also from cross section and fission yield uncertainties. Currently, these uncertainties can't be propagated through burnup calculation with Serpent. Some indication of the uncertainties can be evaluated from Table 9 and the corresponding Figures in Appendix A presenting differences to reference when calculating the nuclide concentrations with different nuclear data libraries. Especially for ^{241}Am and the curium nuclides, differences are large. Differences to all studied nuclide data libraries are several percents with these nuclides. Differences to reference are several percents with all studied nuclide data also for ^{14}C and ^{238}Pu.

Effect of fuel swelling is also not considered in Tables 10 and 11, but according to Table 8 may contribute significantly to uncertainties in the nuclide concentrations. Differences to reference for ^{235}U, ^{242}Pu, ^{242}Cm and ^{244}Cm are between 3 – 5 % when fuel rod gap is assumed to be zero at the end of the irradiation compared to the reference which does not consider fuel swelling. These results indicate that the reference calculation underestimates ^{235}U concentration and taking into account fuel swelling could improve the calculation results with respect to the measured values. However, concentration of ^{242}Pu and the curium nuclides are over estimated by the reference calculation and taking fuel swelling into account would worsen the correspondence to measurements.

Figure 4 presents the uncertainty in the calculated decay heat of sample I2680 (TOT) during the first 300 y and the different uncertainty components contributing to the total uncertainty. In the calculation of the total uncertainty (TOT), the smaller void fraction uncertainty (VOID2) was used. However, the Figure presents also the uncertainty caused by the larger void fraction uncertainty (VOID1) in order to enable comparison. The smaller figure inside Figure 4 zoomes to the first 5 y in order to better see the rapid change in the contributions of power history uncertainty and burnup uncertainty during the first few years of decay.

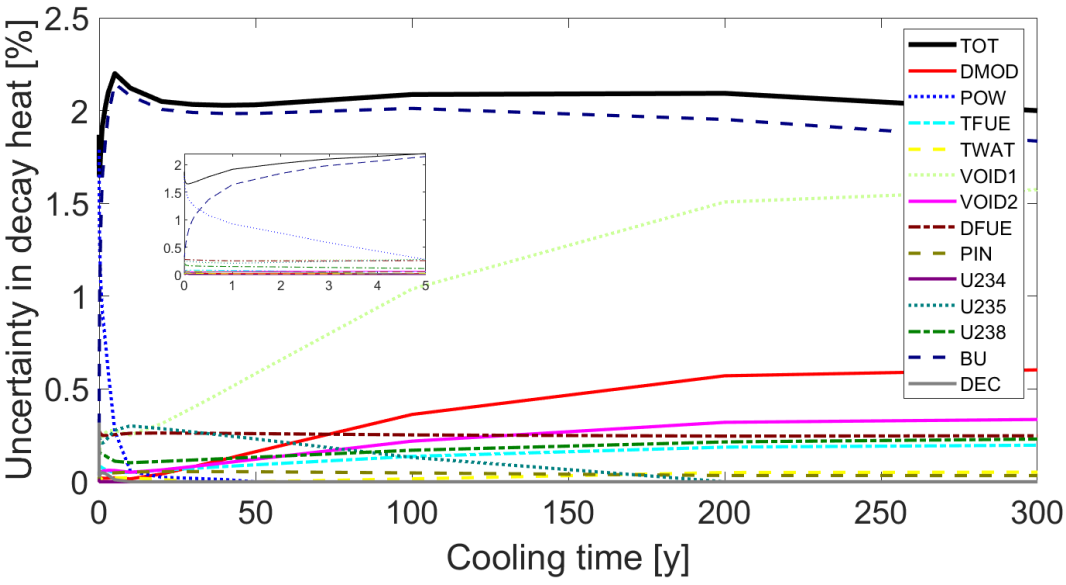

Figure 4. Uncertainty in decay heat calculation. The smaller void fraction uncertainty (VOID2) has been used in the calculation of total uncertainty (TOT).

Even though Table 10 indicates that the most significant source of uncertainty in decay heat determination right after irradiation is power history, Figure 4 shows that the significance of power history quickly dies out. Similar results have been obtained also in another study [14]. Uncertainty in burnup right after irradiation is rather insignifant to decay heat, but also this

changes quickly and burnup becomes the dominant uncertainty source in less than six months after irradiation. If uncertanties in void fraction are large ($\pm$10 % in VOID1), void has a signifant effect to the total uncertainty in decay heat. However, with lower uncertainty in the void fraction ($\pm$2 % in VOID2) the significance of void is approximately of the same order as fuel density, fuel temperature and ^{238}U content and clearly less than 0.4 % within the first 300 y. The effect of moderator density uncertainty somewhat stands out in the figure with maximum 0.6 % uncertainty in decay heat. This suggests that after burnup uncertainty, uncertainty in water density is the second most significant component studied here contributing to the uncertainty in calculated decay heat.

5. Discussion and conclusions

According to the uncertainty analysis, uncertainties in fuel pin radius and ^{234}U enrichment have a very small or insignificant effect on decay heat and all the studied nuclide concentrations at 0 cooling time. Generally, uncertainties in burnup have the largest effect on the calculated quantities. Uncertainties in void fraction may also have a significant effect on many of the concentrations of many of the studied nuclides if void fraction uncertainty is considered large ($\pm$10 %). Even if void fraction uncertainty is considered smaller ($\pm$2 %), it has a clear effect on the concentration of the curium isotopes, many of the plutonium isotopes, ^{14}C and ^{36}Cl. Effects of uncertainties in power density are insignificant apart from ^{238}Pu, ^{242}Cm and decay heat at 0 cooling time. Fuel temperature is only important for some of the heavier nuclides. Only ^{242}Cm and decay heat are sensitive to uncertainties in decay data. It is clear that applying DBRC only on some uranium and plutonium nuclides does not cause uncertainty compared to applying it on all nuclides. However, not applying DBRC at all has a clear $0.5 - 0.6$ % effect on ^{239}Pu and ^{241}Pu concentrations and an effect of $0.1 - 0.4$ % on ^{14}C, ^{36}Cl, ^{235}U, ^{242}Pu and ^{242}Cm concentrations. In the following, decay heat and all studied nuclides are considered separately. For the nuclides, the discussion is limited to the time right after irradiation, but for decay heat the time evolution of uncertainty is also discussed.

At 0 cooling time, **Decay heat** is most sensitive to uncertainties in power density. Uncertainties in moderator density, fuel temperature, water temperature, pin radius and ^{234}U enrichment are insignificant to decay heat at 0 cooling time. Uncertainties in burnup cause only very small uncertainties in decay heat at 0 cooling time. However, in about 2 y after irradiation the uncertainties caused by burnup grow up to around 2 % and remain there for a few hundred years. On the other hand, uncertainties in power density become insignificant within a decade or two which is also evident in Figure 4 in Appendix A and has been observed in another study [14]. Uncertainties caused by moderator density become relatively significant in around 100 y after irradiation when it becomes the second most significant uncertainty component after burnup if smaller void fraction uncertainty ($\pm$2 %) is used. With large uncertainties in void fraction ($\pm$10 %), void fraction is a significant contributor to decay heat uncertainty with greater than $\sim$ 50 y cooling times. Overall, these suggest that uncertainties in water density are not to be neglected when considering decay heat uncertainties in a BWR assembly. Uncertainties in decay data have some ($\sim$ 0.3 %) effect on decay heat uncertainty right after irradiation but become insignificant almost within one day of cooling. Applying DBRC in the calculations has no effect on decay heat up to arounf 10 years of cooling. After 10 years, not using DBRC at all would cause some uncertainty to decay heat. This uncertainty is roughly 0.4 % after 100 years of cooling and smaller before that. According to the sensitivity calculations with different nuclide data, uncertainties due to fission yield and cross sections may have a non-negligible

contribution to decay heat uncertainty. Fuel swelling may cause some smallish uncertainty to decay heat.

Burnup uncertainty is the most significant component in the uncertainties of **^{14}C** and **^{36}Cl** concentrations. Moderator density and void fraction when considering larger void fraction uncertainty also have a significant contribution to the total uncertainty. Power density, fuel temperature, pin radius, ^{234}U enrichment, ^{238}U content and decay data uncertainties have very small or insignificant effect. According to the sensitivity calculations, fuel swelling and nuclear data uncertainties may also have a significant impact on the concentrations of these nuclides.

The fission products **^{137}Cs** and **^{148}Nd** are most sensitive to uncertainties in burnup. Uncertainties in power density, fuel temperature, water temperature, void fraction, pin radius, ^{234}U enrichment and decay data are very small or negligible. Fuel swelling may cause some non-negligible uncertainty to the concentration of these nuclides. Impact of nuclear data uncertainties other than decay data can not be very well predicted from the sensitivity calculations. The same is true for the uranium and plutonium nuclides other than ^{238}Pu. This is because the differences between different nuclear data are not striking.

^{235}U concentration is most sensitive to burnup uncertainty. Significant uncertainty is also caused by void fraction when assuming larger uncertainties, but even with the smaller void fraction uncertainties, this component can't be ignored. ^{235}U is also rather sensitive to moderator density, fuel density and naturally ^{235}U enrichment. Uncertainties in power density, fuel temperature, pin radius, ^{234}U and decay data have very small or negligible effect on ^{235}U concentration. According to the sensitivity calculations, swelling may have a significant contribution to the uncertainty.

Burnup and ^{235}U enrichment are the largest uncertainty components in **^{236}U** concentration. Other rather significant component is fuel density. Power density, fuel temperature, water temperature, void fraction (smaller uncertainty), pin radius ^{234}U enrichment, ^{238}U concent and decay data have very small or negligible effects on ^{236}U concentration.

^{238}U concentration is not very sensitive to any of the examined uncertainty components. Unlike most of the other nuclides, burnup uncertainty has only a very small 0.04 % impact on ^{238}U concentration. Most of the uncertainties are directly caused by uncertainties in ^{238}U concent. Other significant uncertainty source is fuel density. The other uncertainty sources have rather negligible impact on ^{238}U concentration.

Uncertainties in **^{238}Pu** concentration are almost entirely coming from burnup uncertainty if smaller void fraction uncertainty is considered. It is somewhat sensitive to moderator density, power density, void fraction (even with smaller uncertainty), fuel density, ^{235}U enrichment and slightly on ^{238}U content. Fuel and water temperature, pin radius, ^{234}U and decay data uncertainties have negligible effect on ^{238}Pu concentration. Fuel swelling and other nuclear data may also have non-negligible impact on the concentration.

Larger uncertainties in void fraction are the major source of uncertainty in **^{239}Pu** concentration. If smaller void fraction uncertainty is assumed, the major contributor is moderator density, but void fraction is still significant. Other significant uncertainty contributors are water temperature, fuel density, ^{238}U content and burnup. Uncertainties in power density, pin radius, ^{234}U and ^{235}U enrichment and decay data are negligible. Fuel swelling may also have a non-negligible impact.

^{240}Pu concentration is most sensitive to burnup uncertainty. ^{238}U content, fuel density, water temperature and void fraction if larger uncertainty is assumed have clear impact. Power density, pin radius, ^{234}U and ^{235}U enrichment and decay data have a negligible effect on ^{240}Pu concentration. Fuel swelling may also have a non-negligible impact.

^{241}Pu concentration is most sensitive to uncertainties in burnup and void fraction (larger void uncertainty). Moderator density is also a significant uncertainty contributor. Pin radius, ^{234}U and ^{235}U enrichment and decay data have a negligible effect on ^{241}Pu concentration.

Most of the uncertainty in **^{242}Pu** is due to uncertainty in burnup. uncertainties in power density, void fraction, pin radius, ^{234}U enrichment, ^{238}U and decay data have very small effects on ^{240}Pu concentration. Fuel swelling may also have a signifant impact.

Uncertainties in the concentrations of the **curium isotopes** are mostly from uncertainties in burnup. Void fraction uncertainty has a significant impact if larger uncertainty is considered. pin radius and ^{234}U and ^{235}U enrichment have a negligible effect on ^{242}Cm concentration. Power density, fuel density, pin radius, ^{234}U enrichment, ^{238}U content and decay data uncertainty have a negligible impact on ^{244}Cm concentration. According to the sensitivity calculations, fuel swelling and nuclear data uncertainties may have a significant impact on the concentration of both ^{242}Cm and ^{244}Cm.

Small uncertainties ($\lesssim$ 0.1 %) in Table 10 for decay heat and ^{238}Pu and ^{244}Cm concentrations may be questionable due to rather large Monte Carlo variations (standard deviations 0.2 – 0.6 %). For the other plutonium and curium nuclides, standard deviations of the repetition calculations were also 0.1 – 0.2 %. Better accuracy would be achieved using more neutron histories in the calculations. However, this would considerably increase the calculation time.

The better correspondence of the 3D calculation with measurement results for transuranium nuclides is likely related to more realistic neutron spectrum as a function of elevation. The water densities at sample position in the 3D model were slightly smaller than in the 2D model hardening the neutron spectrum relative to the 2D model. Harder spectrum generally leads to greater generation of actinides heavier than uranium. Since both calculations underestimated the concentration of actinides (except ^{238}U) greater concentration of actinides in the calculations improves the correspondence to measurement results.

Acknowledgements

This work has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement N°847593.

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